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Penning-trap Q -value determination of the $^{71}\text{Ga}(\nu, e^-)^{71}\text{Ge}$ reaction using threshold charge breeding of on-line produced isotopes

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ABSTRACT

We present a first direct Q -value measurement of the $^{71}\text{Ga}(\nu, e^-)^{71}\text{Ge}$ reaction using the TITAN mass-measurement facility at ISAC/TRIUMF. The measurements were performed in a Penning trap on neon-like $^{71}\text{Ga}^{21+}$ and $^{71}\text{Ge}^{22+}$ using isobar separation of the on-line produced mother and daughter nuclei through threshold charge breeding in an electron-beam ion trap. In addition, isoionic samples of $^{71}\text{Ga}^{21+}$ and $^{71}\text{Ge}^{21+}$ were stored concurrently in the Penning trap and provided a separate Q -value measurement. Both independent measurements result in a combined Q -value of 233.5 ± 1.2 keV, which is in agreement with the previously accepted Q -value for the ν cross-section calculations. Together with a recent measurement of the ν -response from the excited states in ^{71}Ge , we conclude that there are no further uncertainties in the nuclear structure, which could remove the persistent discrepancy between the SAGE and GALLEX calibration measurements performed with neutrinos from reactor-produced ^{51}Cr and ^{37}Ar sources and the theoretical expectation.

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1. Introduction

As neutrino physics experiments advance to increased precision, there has been a renewed interest in scrutinizing the discrepancy observed in the SAGE and GALLEX neutrino calibration measurements. SAGE and GALLEX have been the two leading initiatives to determine the electron-neutrino flux from the basic solar pp -fusion process [1–5] and thereby significantly contributed to our

modern understanding of neutrino oscillations and neutrino matter effects in the Sun. Both experiments used the $^{71}\text{Ga}(\nu, e^-)^{71}\text{Ge}$ charged-current (CC) reaction and the subsequent 11.43 d electron-capture (EC) decay of ^{71}Ge as a signature for the neutrino reaction. Detector-calibration measurements were subsequently performed with neutrinos from the decays of reactor-produced ^{51}Cr and ^{37}Ar sources [2–6]. Based on neutrino cross-section calculations performed by Bahcall [7], the ratio of measured to predicted ^{71}Ge event rates, however, turned out to be consistently below expectation with an average deficit of 13% and $\approx 2.5 \sigma$ significance according to Ref. [4].

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While arguments are made to explain the deficit with new physics and possible extensions to presently known neutrino properties [8–10], one obvious check is to verify the underlying nuclear physics assumptions as suggested by Haxton [11] and Bahcall [7]. In this context the contributions from the two lowest lying states in ^{71}Ge at 175.0 keV ($5/2^-$) and 499.9 keV ($3/2^-$), which can be reached by the CC reaction from the ^{51}Cr and ^{37}Ar neutrino sources, were recently re-evaluated from a high-precision $^{71}\text{Ga}(^3\text{He}, t)$ charge-exchange experiment to be $7.2 \pm 2.0\%$ [12] for the ^{51}Cr source. This value even slightly amplifies the afore-mentioned SAGE/GALLEX discrepancy, since Bahcall's calculations [7] used in the SAGE/GALLEX calibration were based on a 5.1% contribution.

In view of the importance centered around neutrino physics, a critical review of the various other quantities, which enter into the neutrino cross-section calculation, may therefore be in order. Whereas the decay properties of the ^{51}Cr nucleus are known to rather high precision, we find that there is a need to re-examine the experimental Q -value of the ^{71}Ge EC-decay. One may note that the Q -value enters quadratically in first order into the cross-section calculation, and an 8 keV increase is sufficient to reach consistency at a 1σ -level. Past measurements of the ^{71}Ge Q -value were performed by extrapolating a measured internal bremsstrahlung (IB) spectrum to the end-point energy. The bremsstrahlung component by itself is a highly suppressed process, and photon-emission probability near the end-point drops even further by several orders of magnitude, thereby making precise end-point measurements highly non-trivial. One may also note that the Q -value determination for ^{71}Ge was mostly a side-product of experiments motivated by searches for a 17 keV neutrino [13–15]. The quoted values range from 225 keV to 237 keV [16–22] with $Q = 232.69 \pm 0.15$ keV being the value used by Bahcall [7]. Only one additional Q -value measurement based on a different technique has been reported [23], where a (p, n) charge-exchange threshold measurement gave a Q -value of 236 ± 2 keV.

High-precision Penning-trap mass spectrometry [24,25] is at present the most competitive technique to assess whether the previous ^{71}Ge EC Q -value measurements could have suffered from unknown systematic effects. Many Penning-trap measurements with both singly and highly charged ions (HCIs) [26], using on-line [27,28] as well as off-line [29–36] produced ion beams, have led to numerous Q -values with precisions of order 1 ppb. In the present work we further introduce a new scheme, by which an additional efficient isobaric ion separation can be realized. This separation is based on threshold charge breeding using an electron-beam ion trap charge breeder (EBIT) [37].

2. Experiment and results

The measurement was performed at the ISAC facility at TRIUMF using the TITAN (TRIUMF's Ion Trap for Atomic and Nuclear science) ion trap and mass-measuring setup [38], whose successful operation on unstable nuclei has been demonstrated in a series of recent experiments [39–45].

TITAN consists of three essential components, a radio-frequency quadrupole cooler and buncher (RFQ) [46], the afore-mentioned EBIT, and a precision mass-measurement Penning trap (MPET) [39].

For the present measurement the two isotope species, ^{71}Ge and ^{71}Ga , were co-produced by irradiating Ta target foils with a 50 μA proton beam at 500 MeV from the TRIUMF main cyclotron. The TRIUMF Resonant Ionization Laser Ion Source (TRILIS) [47] provided a beam of resonantly ionized Ge atoms in parallel to surface-ionized Ga atoms. One may note that germanium does not easily surface ionize due to its larger ionization potential of

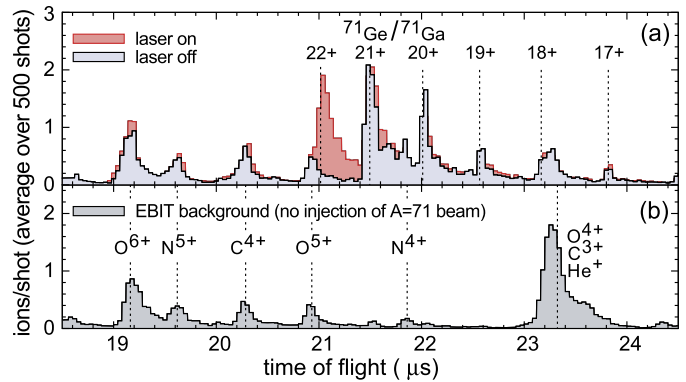


Fig. 1. Time-of-flight spectra of ions extracted from the EBIT at a transport energy of ≈ 1.5 keV $\times q$. The electron-beam energy of 2.00 keV ionizes Ge and Ga up to neon-like charge states. (a) Comparison of spectra from a mono-isotopic Ga (laser off) and from a Ga/Ge (laser on) mixed beam injection. The peak at 21 μs corresponds to ions with $m/q = (71/22)$ and appears, if the laser is on. (b) Background spectrum from residual gas, which is charge bred in the EBIT also when the $A = 71$ beam injection is turned off.

$E_{\text{IP}} = 7.9$ eV as opposed to 6.0 eV for gallium [48]. The titanium-sapphire laser-ionization scheme, which was developed off-line by Kessler et al. [49], uses a 3-step resonant excitation into an auto-ionizing state of germanium. This scheme was successfully employed on-line for the first time and provided a $^{71}\text{Ge}^+$ beam intensity of $1.8 \cdot 10^7$ ions per second, as measured at the ISAC yield station. It allowed switching from a mono-isotopic ^{71}Ga to a mixed $^{71}\text{Ga}/^{71}\text{Ge}$ ion beam by blocking or unblocking the laser (cf. Fig. 1).

The ions were extracted as 20 keV ion beams, mass separated and delivered to the TITAN RFQ, where they were accumulated, cooled and bunched. The bunches from the RFQ were subsequently captured in the EBIT [50] for fast charge breeding through successive electron-impact ionization. The electron beam was set to a current of 70 mA and an energy of 2.00 keV, which is just at the ionization-potential thresholds of neon-like ions, i.e., $E_{\text{IP}}(\text{Ga}^{21+}) = 2.01$ keV and $E_{\text{IP}}(\text{Ge}^{22+}) = 2.18$ keV [48]. Therefore, for germanium one can reach the charge state $q = 22$, whereas for gallium only $q = 21$. This allows subsequent separation of the two ion species owing to the different m/q ratios. It is the first time that this threshold charge-breeding scheme was applied to separate isobaric samples of on-line produced isotopes.

Fig. 1(a) displays the time-of-flight spectra of ion bunches extracted from the EBIT and detected on a micro-channel plate detector located in front of the MPET. For comparison, the background spectrum (from the residual gas) extracted with the $A = 71$ beam injection turned off is plotted in Fig. 1(b). The EBIT extraction parameters were chosen such that some fraction of the ions remained in the trap for continued charge breeding [50]. This accumulation enabled delivery of HCIs at time intervals shorter than the actual charge-breeding time, and it produced a charge-state distribution, which peaked at the atomic shell closure. Close to 30% of the extracted Ga or Ge ions appeared in the Ne-like charge state. With the resonant laser-ionization scheme for Ge active, a mono-isotopic Ge^{22+} beam was delivered to the MPET, and a mono-isotopic Ga^{21+} beam was achieved with the laser off. A fast ion gate of the Bradbury–Nielsen (BN) type [51] allowed q/m selection for the mass measurement.

In the MPET the cyclotron frequency $f_c = \frac{e}{2\pi} \frac{q}{m} B$, and hence the mass m , was determined using the time-of-flight ion-cyclotron-resonance method (TOF-ICR) [52,26]. With this technique the mass resolution at a given magnetic field B is inversely proportional to the radio-frequency excitation time T_{rf} and to q , i.e.,

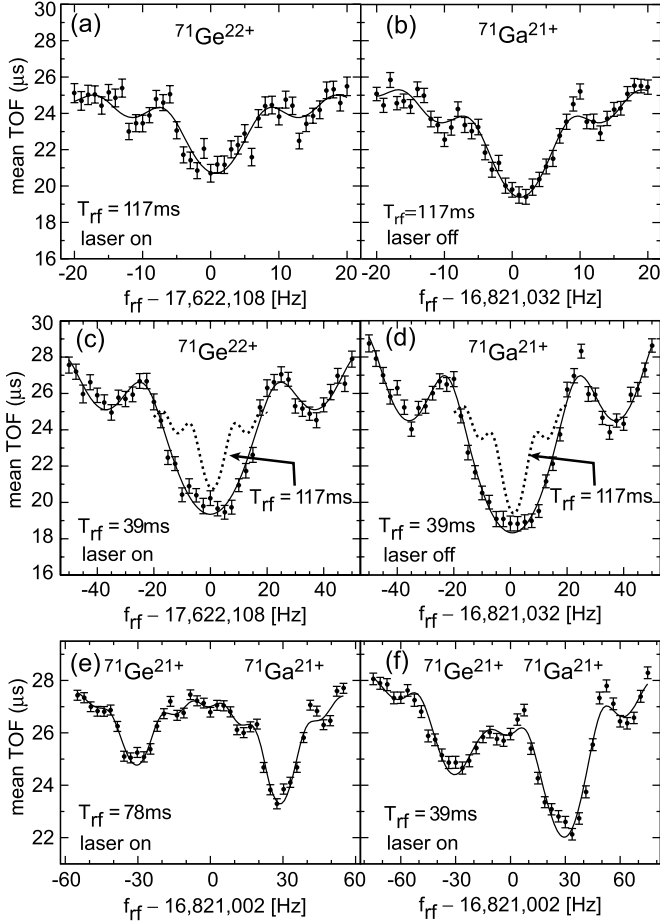


Fig. 2. TOF-ICR spectra for the mono-isotopic $^{71}\text{Ge}^{22+}$ (a, c), $^{71}\text{Ga}^{21+}$ (b, d) and the isobaric two-component beams (e, f) recorded with different rf-excitation times T_{rf} (note the different frequency scales). Solid lines are line-shape fits to the data [52]. Longer excitations result in narrower resonances, however, at the cost of increased charge-exchange and damping effects. This is exemplified by comparing the curves of (a, b) (dashed lines) with the ones in (c, d). The resonances for the $q = 21$ isobaric two-component beams are displayed in (e, f) for $T_{\text{rf}} = 78$ ms and $T_{\text{rf}} = 39$ ms showing a similar effect on T_{rf} .

$$\frac{\Delta m}{m} \propto \frac{m}{eqBT_{\text{rf}}}, \quad (1)$$

which makes the use of highly charged ions particularly interesting. Alternating measurements on Ga^{21+} and Ge^{22+} of typically 13 min acquisition time were performed, and throughout the campaign different settings were chosen for the excitation time and the magnetron radius (controlled by the Lorentz-steering potential U'_{LS} [53]), in order to study systematic effects like, e.g., the interaction of HCl with the residual gas. Typical TOF-ICR spectra with different T_{rf} are shown in Fig. 2. Charge-exchange and damping effects diminish the quality of the TOF-ICR data for longer T_{rf} (i.e. trapping time in the MPET), however, no systematic shifts were observed. Furthermore, an O^{5+} contamination to the $m/q = (71/22)$ beam could not be entirely removed by the BN-gate. It explains the slightly less pronounced resonance for the $^{71}\text{Ge}^{22+}$ ions compared to $^{71}\text{Ga}^{21+}$ (cf. Fig. 2(a)–(d)). However, the contamination was less than 20% and was well separated in excitation frequency at the MPET ($\Delta\nu = 137,072$ Hz). A count-class analysis did not reveal any significant shifts. This contamination may therefore be excluded from being a source of error.

The laser ionization resulted in ^{71}Ge yields comparable to those of surface-ionized ^{71}Ga . This allowed an independent determination of the Q -value by concurrently injecting both elements

Table 1

List of contributing errors of the two independent TITAN Penning-trap Q -value measurements in units of keV.

Contribution	Mono-beam	Mixed beam
σ_{stat} without count-class ¹	0.20	0.51
σ_{stat} with count-class	0.30	0.71
Long-term stability	$< 1\sigma_{\text{stat}}$	n/a
Relativistic shifts ²	$< 1\sigma_{\text{stat}}$	n/a
T_{rf} and ion-ion interactions	$< 1\sigma_{\text{stat}}$	1.71
m/q shifts (trap compensation) ³	1.50	n/a
σ_{tot}	1.53	1.85

¹ Not employed in the final analysis.

² Evaluated using $^{14}\text{N}^{4+}$ and $^{16}\text{O}^{5+}$ ions.

³ Deduced on-line with $^{84}\text{Kr}^{25+,26+}$, off-line with $^{14}\text{N}^{4+}/^{16}\text{O}^{5+}$.

isoionic at charge state $q = 21$ into the MPET. Cyclotron frequencies for both species could be obtained from one TOF-ICR spectrum. Examples of this “double-resonance” are shown in Fig. 2(e)–(f) for $T_{\text{rf}} = 78$ ms and 39 ms, respectively. The signal-to-noise ratio of this method is intrinsically lower than in the case of a mono-isotopic beam leading to larger statistical uncertainties for the same acquisition time. The method is also more sensitive to the modulation accuracy of the analogue rf-ramping. However, systematic shifts resulting from any time dependent fluctuations, relativistic shifts, and/or different m/q ratios are largely eliminated by this simultaneous isoionic measurement.

For both methods the Q -value is calculated from the ratios of the measured frequencies and the charge states

$$R = \frac{f_c^{\text{Ga}}}{f_c^{\text{Ge}}} \cdot \frac{q^{\text{Ge}}}{q^{\text{Ga}}} \quad (2)$$

as

$$Q = M^{\text{Ga}}(R - 1) - m_e(Rq^{\text{Ga}} - q^{\text{Ge}}) + RB^{q^{\text{Ga}}} - B^{q^{\text{Ge}}}, \quad (3)$$

where M^{Ga} is the atomic mass of ^{71}Ga , m_e the electron mass, and $B^{q^{\text{Ga}}}$, $B^{q^{\text{Ge}}}$ are the ionization energies, i.e., the sum of all individual electron-binding energies from the neutral atom towards the highly charged ion at charge state q . Electron-binding energies were taken from Ref. [54], which have estimated uncertainties of order ± 30 eV in the relevant q -range.

We note that the use of HCl increases the ion-ion interaction in the trap causing frequency shifts [55]. The effect can be accounted for through a count-class analysis, as described in Ref. [56], however, at the cost of slightly increased statistical uncertainties (cf. Table 1).

Consecutive runs of Ga^{21+} and Ge^{22+} have been interleaved with measurements on $^{84}\text{Kr}^{25+,26+}$ and systematic shifts of up to 1.5 keV between the two charge states of Kr have been observed. Therefore, high-statistic accuracy checks using $^{14}\text{N}^{4+}$ and $^{16}\text{O}^{5+}$ were performed after the on-line campaign and confirmed systematic effects on the order of 1.5 keV. The trap compensation settings were deduced from previous comprehensive studies on singly charged ^6Li and ^7Li [39], where sub-ppb precision was achieved. However, these settings were not optimal for HCl in the m/q range of this study, and we cannot exclude systematic uncertainties on the order of 20 ppb for the results presented here. The summary of measurements is shown in Fig. 3(a).

For the $^{71}\text{Ga}/^{71}\text{Ge}$ isobaric two-component beam measurement the average number of ions injected into the MPET was varied and two different T_{rf} were chosen. These results are shown in Fig. 3(b). The count-class analysis revealed systematic shifts of f_c of 0.42 Hz per additional detected ion (equivalent to 25 ppb) for both isobars. As both resonances shifted by a similar extent in the same direction, no significant trend for the derived Q -value was found.

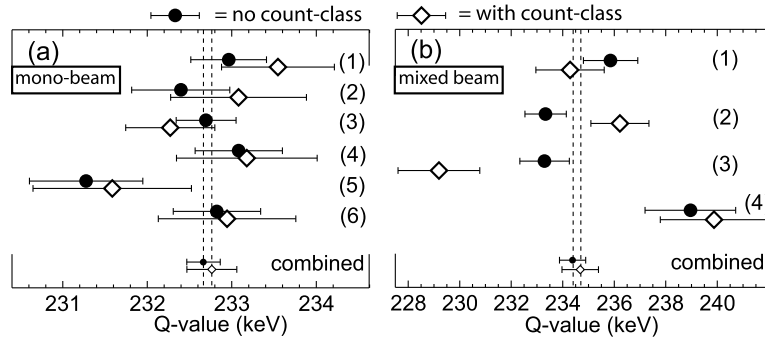


Fig. 3. Summary of Q -value measurements. Figure (a) shows six data sets from a series of measurements with mono-isotopic beams of Ga^{21+} and Ge^{22+} and figure (b) shows four data sets from a series of measurements with a mixed beam of Ga and Ge in charge state $q = 21$. Diamonds/dots show the results with/without count-class analysis [56] together with their combined averages (also indicated by the broken lines). The various numbered sets in (a) indicate measurements with different excitation times T_{rf} and different magnetron radii resulting from different Lorentz-steering potentials U'_{LS} [53], which are for T_{rf} and U'_{LS} , respectively: (1) 78 ms, 50 V; (2) 156 ms, 50 V; (3) 117 ms, 50 V; (4) 117 ms, 60 V; (5) 117 ms, 30 V; (6) 39 ms, 50 V. The average number of ions detected in all cases was close to one. The sets in (b) differ in excitation times and average number of ions N_{ion} in the trap, which are: (1) 78 ms, 2.0; (2) 78 ms, 2.4; (3) 39 ms, 3.0; (4) 78 ms, 3.5. In these cases the Lorentz-steering potential was kept constant at 50 V.

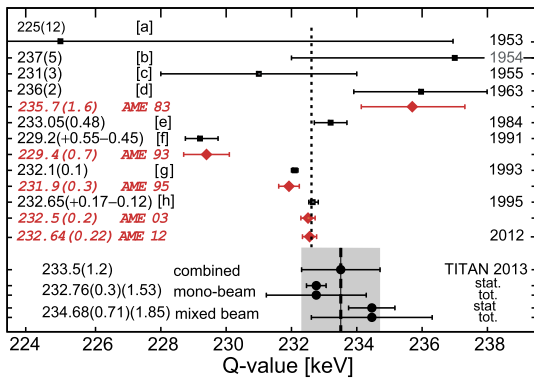


Fig. 4. Comparison of Q -value measurements (1σ errors) ([a] = Ref. [16], [b] = Ref. [17], [c] = Ref. [18], [d] = Ref. [23], [e] = Ref. [19], [f] = Ref. [20], [g] = Ref. [21], [h] = Ref. [22]) leading to various updated AME values (diamonds) in a given year [58–62]. The individual IB measurements scatter by up to 6.5σ . The dotted line represents the value taken by Bahcall [7]. The Q -value measurements from this work (circles) are shown for comparison with the average value indicated by the dashed line and the error indicated by the shaded area.

Without count-class analysis the scatter of the Q -values for different settings of T_{rf} and average number of ions resulted in a Birge ratio [57] (which is a measure of the appropriateness of statistical uncertainties) of 1.74, and by including count-class analysis it increased to 2.60. This could point to possible ion–ion interaction effects so far unaccounted for. The total uncertainty was therefore conservatively evaluated to 1.85 keV (in accordance with the Birge ratio of 2.60, or with a systematic uncertainty of 1.71 keV added in quadrature to the statistical uncertainty of 0.71 keV, cf. Table 1). A more comprehensive discussion of the data analysis will follow in a forthcoming publication.

Our results are compared to indirect IB end-point measurements [17–22] and AME values [58–62] as shown in Fig. 4. The various error contributions are listed in Table 1. The two independent mass-measurement methods as described in this work agree within their total uncertainties, and they result in a weighted average Q -value of $Q = 233.5 \pm 1.2$ keV. This value does not change the calculated ^{51}Cr ν cross section to the extent as to reduce the observed GALLEX and SAGE discrepancy in a significant way.

3. Conclusion

In summary, the Q -value of the $^{71}\text{Ga}(\nu, e^-)^{71}\text{Ge}$ reaction has been determined in a direct Penning-trap measurement using

HCLs, whereby on-line co-produced isobars have been separated through threshold charge breeding for the first time. The TITAN result firmly excludes an incorrect Q -value as a cause for the gallium anomaly observed in the GALLEX and SAGE calibration runs. Combined with the recent ν -response measurements reported in Ref. [12], we conclude that there are no further uncertainties in the nuclear structure, which could remove the discrepancy.

Charge breeding around thresholds of electron-shell closures can provide clean mono-isotopic ion beams, and thus opens up new opportunities for radioactive isotope facilities beyond those of increasing the mass resolution in Penning-trap measurements. Especially on the proton-rich side, where intense contaminations from less proton-rich isobars are frequently present in ion beams at ISOL-type facilities, the technique described can be a valuable advantage.

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